

Magnetoresistive memory in phase separated $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$

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We have studied a non volatile memory effect in the mixed valent compound $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ induced by magnetic field (H). In a previous work [R.S. Freitas et al., Phys. Rev. B 65 (2002) 104403], it has been shown that the response of this system upon application of H strongly depends on the temperature range, related to three well differentiated regimes of phase separation occurring below 220 K. In this work we compare memory capabilities of the compound, determined following two different experimental procedures for applying H, namely zero field cooling and field cooling the sample. These results are analyzed and discussed within the scenario of phase separation.

Keywords: Manganites, phase separation, memory

Rare earth based manganese oxides, also known as manganites, have been the focus of extensive research since the discovery of the colossal magnetoresistance effect (CMR) [1]. The close interplay between ferromagnetic double exchange [2] and antiferromagnetic superexchange gives rise to extraordinary properties. The most intriguing one is the existence of a phase separated state, i.e. the coexistence of ferromagnetic metallic (FM) and charge ordered (CO) insulating phases [3].

The possibility to manipulate the relative fraction of the coexisting phases has been widely studied using several techniques. Phase separation can be altered by introducing chemical disorder [4], by changing the ceramic grain size of the samples [5, 6], by applying external hydrostatic pressure [7], by the application of external electric [8] and magnetic fields [9, 10] and by thermal cycles of the samples passing through a first order phase transition [11].

In previous works [12, 13] we have explored the possibility of controlling the relative amount of the coexisting phases in $\text{La}_{5/8-x}\text{Pr}_x\text{Ca}_{3/8}\text{MnO}_3$ ($x=0.3$, LPCMO) and $\text{La}_{0.5}\text{Ca}_{0.5}\text{Mn}_{1-y}\text{Fe}_y\text{O}_3$ (LCMFO) by the application of magnetic field.

$\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ (LCMO) is one of the most studied system in the literature of manganite materials. The compound, paramagnetic at room temperature, changes on cooling to a mainly FM metallic phase at $T_C \approx 220$ K, and subsequently to charge-ordered antiferromagnetic (CO-AFM) phase at $T_{co} \approx 150$ K (180 K upon warming)[14]. However, it has been established that this system is better described as magnetically phase-segregated over a wide range of temperatures [5, 15], a phenomenon called phase separation (PS). At low temperatures, $T < T_{co}$, FM metallic regions are trapped in a CO-AFM matrix, whereas at an intermediate temper-

ature range ($T_{co} < T < T_C$), the FM phase coexists with insulating non-FM regions.

It was established that three ranges can be identified in which this phase coexistence exhibits different features under the application of H [15]:

- A soft PS state for $200 \text{ K} < T < 220 \text{ K}$: FM clusters coexist with paramagnetic regions. In this range, ρ is reversible against the application of moderate magnetic fields.

- An intermediate PS for $150 \text{ K} < T < 200 \text{ K}$: Coexistence of FM and insulator regions. The FM phase is partially confined but can grow against the insulating one while applying a low H.

- A hard PS state for $T < 150 \text{ K}$: Coexistence of FM and CO-AFM regions. The FM phase is structurally confined and cannot grow against CO in moderate H.

In this report we have studied the possibility of imprinting different values of H by inducing the irreversible growth of FM and CO regions by applying magnetic fields in the field cooling (FC) and zero field cooling (ZFC) modes in different temperature ranges. Their relative amount acts as a sort of analogical memory of the previously applied H. To recover the H value, M or ρ measurements can be used.

Polycrystalline samples of $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ were used, their preparation procedure and structural characterization are described elsewhere [5]. Transport measurements were performed with the standard four probe method. Magnetization measurements were performed in a commercial magnetometer (Quantum Design PPMS).

Measurements were made using the the ZFC and the FC procedures:

In the ZFC mode the sample is cooled to the desired T in a field $H_0 = 0$ for ρ measurements. For M measurements a ZFC-like procedure was performed cooling in a small $H_0 \approx 0.1 \text{ T}$. Then, different $H_{ap} (> H_0)$ are turned

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on and off during periods of tens of minutes, increasing H_{ap} on each run. We expect to write the different values of H_{ap} , in the relative growth of the FM phase. In this case, H_{ap} is the perturbation.

Measurements in the ZFC mode were performed in the 150 K - 200 K range, where we expect an irreversible change of the properties [15].

Figure 1 (a) and (b) shows M and the applied H vs time and ρ and the applied H vs. time respectively, for experiments performed in the ZFC procedure at 170 K.

Sudden increases of M and decreases of ρ are observed when H_{ap} is applied. These jumps are similar to the ones observed in LPCMO and LCMFO [12, 13] and are related to the (fast) alignment of spins and domains and to the (slow) enlargement of the FM phase. Remarkably, once H_{ap} is turned off, M decreases and ρ increases without recovering their previous H_0 values. Thus, the presence of a persistent effect directly related to the magnitude of the previously H_{ap} is apparent.

This effect was found in the 150 K - 200 K temperature range, while below T_{co} it is almost negligible.

In agreement with previous results on LPCMO and LCMFO [12, 13], we can relate this effect to the increase of the FM fraction (f) of the sample every time an increasing H pulse is applied. However, we have to note that the effect is less significant in the present work than the one observed in the above mentioned references.

By using M measurements, the relative change of f after the whole experiment and the application of $H = 6$ T can be estimated by the ratio of the "initial" and "final" values of magnetization under no perturbation, indicated in the figure as M_i and M_F . With these values, we obtain $f_F/f_i \approx 1.10$, i.e. an increase of 10 % in the FM fraction.

Assuming a linear dependence of Δf with H , this would mean an increase of less than 2 % of f by Tesla. Also, from fig. 1(b), we can estimate $MR / H \approx 5\%/Tesla$, a very low value when compared to the almost 80%/Tesla achieved by LPCMO [12].

As LCMO exhibits robust CO features in a broad T range, we have explored an alternative way to produce memory effects, inducing the growth of CO regions by field cooling the sample (FC mode).

In the FC mode the sample is cooled to the desired T in $H_{FC} \approx 1$ T. We have measured ρ and M while applying short "negative" field pulses, i.e. reducing the magnetic field to $H_{ap} < H_{FC}$ and returning after each pulse to H_{FC} . The perturbation in this procedure is given by pulses of $\Delta H = H_{FC} - H_{ap}$. In this case, we expect the compound to memorize the applied ΔH value in the growth of non - FM regions.

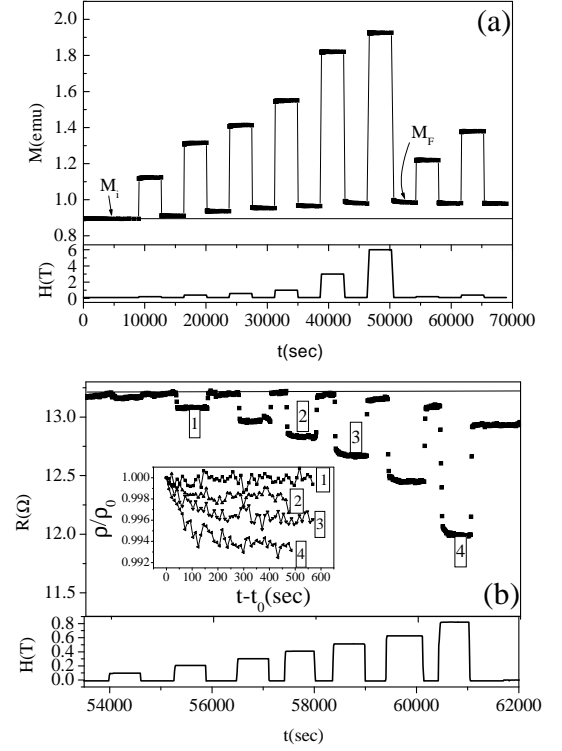


FIG. 1: ZFC procedure: (a) $M(170K)$ vs time for LCMO upon the application of $H = 0.2, 0.4, 0.6, 1, 3, 6, 0.2$ and 0.4 T over H_0 . (b) $R(170 K)$ vs time for LCMO upon the application of $H = 0.1, 0.2, 0.3, 0.4, 0.5, 0.6$ and 0.7 T. Inset: normalized $\rho(170 K)$ vs time during the onset of H . Number labels correspond to the ones in main figure.

FC measurements were performed below T_{co} , where the CO is harder.

Experiments performed at $T = 130$ K are shown in fig. 2 (a) and (b), where we see that a much larger effect can be obtained by making this simple modification of the ZFC experimental procedure. In the ZFC mode a change of around 10 % was observed in f as mentioned above, while for the FC mode, a change of almost 40 % can be achieved after the application of $\Delta H \approx 2$ T as can be obtained from fig. 2.

The application of the field pulses in the FC procedure to the sample (see fig. 2), result in a huge reduction of the M value and an equivalent increase of ρ , after returning to H_{FC} .

The change observed in this last experiment is related to the increase of the CO phase during the temporary reduction of the field.

The applied H_{FC} forces a FM state in regions that otherwise would be CO. The subsequent return to H_{FC} , yields an effect similar to that obtained for the FM regions in the ZFC procedure.

The inset of fig. 2(b) shows the relaxation of the

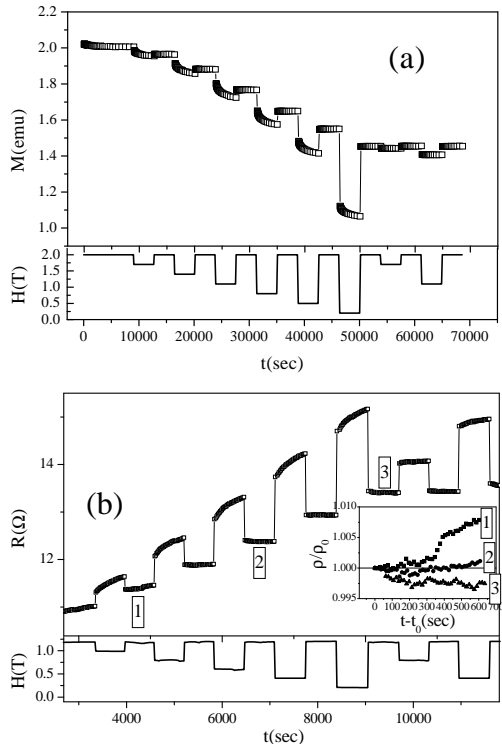


FIG. 2: FC procedure: (a) $M(130K)$ vs time for LCMO upon the reduction of H in 0.3, 0.6, 0.9, 1.2, 1.5, 1.8, 0.3 and 0.9 T ($H_{FC} = 2$ T). (b) $R(130 K)$ vs time for LCMO upon the reduction of H in 0.1, 0.2, 0.3, 0.4, 0.5, 0.2 and 0.4 T ($H_{FC} = 0.7$ T). Inset: normalized $\rho(130 K)$ vs time after H has returned to H_{FC} . Number labels correspond to the ones in main figure.

system after the field returns to H_{FC} . This relaxation cannot be seen on M measurements due to the relatively small magnitude of the changes obtained, but is clear on resistivity ones. After reducing the field in $\Delta H = 0.1$ T (labeled as relax. 1), relaxation is towards an increment in ρ . After $\Delta H = 0.2$ T (labeled as relax. 2), the system do not show relaxation and on subsequent increases of ΔH , ρ reduces with time. This last result indicates the critical value ΔH_{crit} above which the CO phase attains an overenlarged metastable state, and signs the existence of an equilibrium value of the FM (or CO) fraction which depends on H_{FC} .

As a common feature for both procedures, we can observe that the memory effect related to changes in the relative fraction of the phases, is persistent after the "perturbation" (increase in H in the ZFC mode, decrease in H in the FC mode) is turned off, and can only be modified if an ulterior higher perturbation is applied.

In both procedures we can see that alignment and enlargement effects are present. We argue that the enlargement of the FM phase in the ZFC mode and of the CO phase in the FC mode is the responsible of the memory effect. Because of this, if we apply a smaller perturbation than the last applied, only alignment effects are expected, as actually happens (see fig. 1 and 2).

Additionally, we can make an estimation of the change in f in the FC procedure using M and ρ measurements.

For M measurements, the same procedure as presented for the ZFC mode can be used. For resistivity measurements, we can make an indirect determination of f using a semi phenomenological model for transport through a binary mixture known as General Effective Medium theory or GEM [16], which provides a relation to obtain the samples' resistivity (ρ_e) as a function of the resistivities of the constitutive phases (see refs. [9, 10]).

From our M measurements, we obtain $\Delta f/H \approx 16\%/Tesla$, while for ρ , the value slightly changes to $14\%/Tesla$.

Summarizing, we have observed a persistent effect that can be used as an analogical memory of previously applied magnetic fields. The effect is related to the enlargement of the FM or the CO phase according to the particular procedure (ZFC or FC). We have shown that temperature is very significant and has to be taken into account when studying this effect. A previous characterization of the system (as in [15] for LCMO) is useful to choose the appropriate temperature range to obtain a more sensitive response of the system. We have found that, for LCMO, FC seems to be the better procedure because larger changes can be observed in both M and ρ for changes in H of the same order of magnitude. We have also observed that this effect is enhanced in the range in which the CO phase is harder (see [15]).

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